Competition between singlet and triplet pairings in $Na_x\ CoO_2 \cdot y\ H_2O$

Kazuhiko Kuroki
Department of Applied Physics and Chemistry, The University of Electro-Communications, Chofu, Tokyo 182-8585, Japan

Yukio Tanaka
Department of Applied Physics, Nagoya University, Nagoya, 464-8603, Japan

Ryotaro Arita
Department of Physics, University of Tokyo, Hongo 7-3-1, Tokyo 113-0033, Japan

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We discuss the pairing symmetry of a cobaltate superconductor $Na_xCoO_2 \cdot y\ H_2O$ by adopting an effective single band model that takes into account the e'_g hole pockets, as discussed in our previous paper [to appear in Phys. Rev. Lett.] Here we consider the off-site repulsions in addition to the on-site repulsion considered in our previous study. We show that the spin-triplet f-wave pairing proposed in our previous study is robust to some extent even in the presence of off-site repulsions. However, f-wave pairing gives way to singlet pairings for sufficiently large values of off-site repulsions. Among the singlet pairings, i-wave and extended s-wave pairings are good candidates which do not break time reversal symmetry below T_c in agreement with the experiments.

I. INTRODUCTION

Discovery of superconductivity in a hydrated sodium cobaltate $Na_xCoO_2{\cdot}yH_2O$[1] has attracted much attention recently. This material is of special interest since it is a $3d$-electron system having a quasi-two-dimensional lattice structure, which resembles the high T_c cuprates. On the other hand, it differs from the cuprates in that the Co ions form a triangular lattice, and also there are several bands intersecting the Fermi level, all of which are away from half-filling. Another interesting point concerning this material is that some experiments suggest possible occurrence of an unconventional superconductivity.[2,3,4,5] In particular, there has been a debate as to whether the pairing occurs in the spin singlet channel or in the triplet channel. An unchanged Knight shift across T_c found in some experiments suggests spin-triplet pairing,[3] while the Knight shift does decrease below T_c in other experiments at low magnetic field.[6,7] As for the occurrence of broken time reversal symmetry, several μSR experiments show that such a possibility is rather small.[8,9]

There has also been a singlet-triplet debate theoretically. Some theories propose singlet pairing,[11,12,13,14], others triplet pairing mechanisms.[15,16,17,18,19,20] Naively, presence of possible ferromagnetic spin fluctuations reported in some experiments[5,6] may support spin-fluctuation-mediated triplet pairing scenario. Couple of years ago, however, two of the present authors showed that T_c of spin triplet superconductivity due to ferromagnetic spin fluctuations, if any, should in general be very low.[21] A similar conclusion has been reached by Monthoux and Lonzarich.[22] This is because the triplet pairing interaction mediated by spin fluctuations is proportional to $\chi/2$ (in contrast to $3\chi/2$ for the singlet case), where χ is the spin susceptibility, while the effective interaction that enters the normal self energy is proportional to $3\chi/2$, so that the suppression of pairing due to the normal self-energy overpowers the pairing effect. We then showed that this difficulty for spin-fluctuation-mediated triplet pairing may be eased in systems having 'disconnected Fermi surfaces', where the nodal lines of the gap can run *in between* the Fermi surfaces to open up a full gap *on the Fermi surfaces*.[23] Since pairing with high angular momentum is degraded by the presence of gap nodes on the Fermi surfaces, opening a full gap may result in an enhanced pairing.[23,24,25,26,27,28] As an example, we considered the Hubbard model on a triangular lattice, where the Fermi surface becomes disconnected into two pieces centered around the K point and the K$'$ point (see Fig.1(a)) when there are a small number of holes (or electrons, depending on the sign of the hopping integral). Using fluctuation exchange (FLEX) approximation[29,30,31] and solving the linearized Éliashberg equation, we have shown that a finite T_c can be obtained for spin-triplet f-wave pairing, where the nodes of the gap run between the Fermi surfaces.[23] We have also confirmed this conclusion[32] using dynamical cluster approximation,[33] which is a non-perturbative approach.

In a recent study,[18] we have looked at the band calculation results of Na_xCoO_2[34] from this point of view, where we find that the pocket-like Fermi surfaces (hole pockets) near the K and K$'$ points (as in the bottom figure of Fig.1(b)), originating from the band having e'_g character to some extent in the notation of ref.34 (denoted by the thick line in Fig.2(a)), are disconnected in a similar sense as in the triangular lattice, namely the nodes of the f-wave gap do not intersect the Fermi surfaces (although they do intersect the large Fermi surface around the Γ point originating from the bands having a_{1g} character).

In fact, a strong motivation to focus on the e'_g hole pockets is the presence of van Hove singularity (vHS) near the K point. Namely, as can be seen from the band structure shown in Fig.2, there exist saddle points at points denoted as SP, where the density of states (DOS) takes a large value. Since this large DOS lies close to the

Fermi level[34], it is likely that the band structure around the K and K′ points strongly affects the low energy properties of this material. In particular, ferromagnetic spin fluctuations may arise due to this high DOS near the Fermi level.

In ref.18, we have adopted a model where we separate out the portion of the bands which has e'_g character to some extent, and in particular focus only on the upper e'_g band, which contributes to the formation of the pocket Fermi surfaces and to the large DOS at the vHS, while neglecting the lower e'_g band, which has only small DOS near the Fermi level due to the linear dispersion at the band top. Although this portion of the band, shown by the thick curves in Fig.2(a), is disconnected between M and Γ points due to a_{1g}-e'_g hybridization, it originally comes from a single band having e'_g character, as can be understood from the inset of Fig.2(a), where a tight binding band dispersion is given for a case when large a_{1g}-e'_g level offset is introduced. We have found that the thick portion of the band in Fig.2(a) (apart from the missing part between M and Γ points) can be roughly reproduced by a single band tightbinding model on a triangular lattice with hopping integrals up to fourth nearest neighbors. Namely, the dispersion of the tight binding model on an isotropic triangular lattice with only nearest neighbor hoppings takes its maximum at the K point (Fig.1(a)), while the band top moves towards the Γ point when the fourth neighbor hopping is introduced (Fig.1(b)), resulting in a pocket like Fermi surface that lies between the K and Γ points when small amount of holes are present. Moreover, the band structure around the K point resembles that of the actual material (compare Fig.1(b) and the thick curve in Fig.2), so the vHS due to the saddle points SP in Fig.2 is also reproduced by this effective model. As for electron-electron interactions, we considered a model with only the on-site repulsion, where we found using FLEX that spin triplet f-wave pairing indeed dominates and has a finite T_c.[18]

In the present study, we continue to study the single band model having hoppings up to fourth nearest neighbors, but this time with repulsive interactions up to second nearest neighbors to investigate the effect of the off-site repulsions. We find that (i) spin triplet f-wave pairing is robust to some extent even in the presence of off-site repulsions, but (ii) for sufficiently large off-site repulsions, singlet pairings can dominate over triplet pairings.

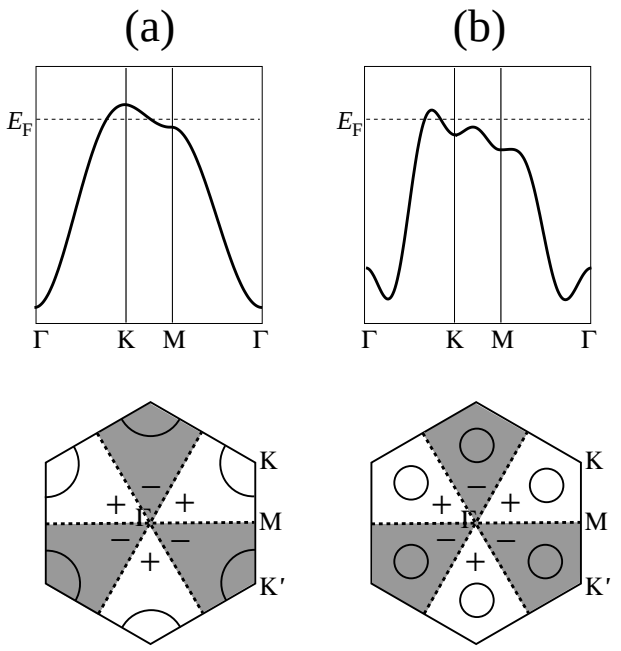

FIG. 1: Band dispersion (top) along with the Fermi surfaces (schematic) for a small number of holes and the f-wave gap (bottom) are shown for a tight binding model on a triangular lattice with (a) only nearest neighbor hopping, and (b) when fourth nearest neighbor hopping $t_4 = 0.2$ is introduced. $+-$ denote the sign of the gap function, while the dashed lines represent the nodal lines.

II. FORMULATION

A. Model

In standard notations, the Hamiltonian considered in the present study is given in the form,

$$H = \sum_{ij,\sigma} \left(t_{ij} c^\dagger_{i\sigma} c_{j\sigma} + \text{h.c.} \right) + U \sum_i n_{i\uparrow} n_{i\downarrow} + V_1 \sum_{\langle ij \rangle', \sigma, \sigma'} n_{i\sigma} n_{j\sigma'} + V_2 \sum_{\langle ij \rangle'', \sigma, \sigma'} n_{i\sigma} n_{j\sigma'} \qquad (1)$$

where $t_{ij} = t_1$, t_2, t_3, and t_4 are the nearest, second nearest, third nearest and fourth nearest neighbor hopping integrals, respectively,[35] and U, V_1, V_2, represent on-site, nearest neighbor, second nearest neighbor electron-electron repulsions, respectively. The summations $\sum_{\langle ij \rangle'}$ and $\sum_{\langle ij \rangle''}$ are taken over pairs of nearest and next nearest neighbors, respectively. We define the hole density as $n_h = 2 - n$, where n is the electron band filling (number of electrons/number of sites). n_h is restricted to a small number in the present study because the e'_g hole pockets are small. We take $-t_1 = 1$ as the unit of the energy throughout the study.

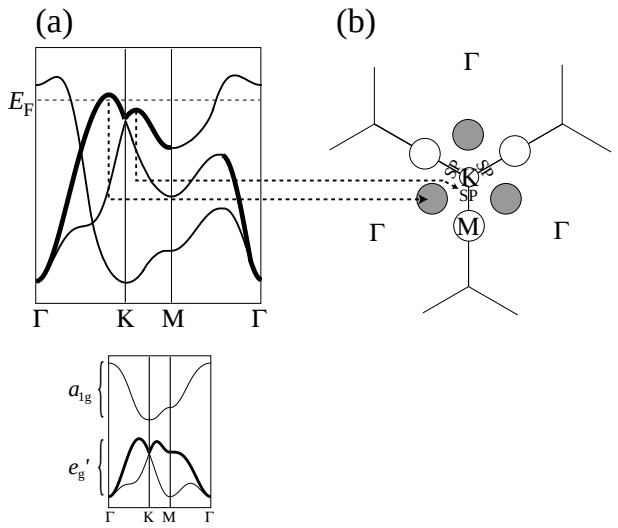

FIG. 2: (a) A schematic plot of the band structure of Na_xCoO_2 following the calculation results of ref.34. The thick line denotes the portion of the upper part of the bands which has e_g' character to some extent. Bottom inset: dispersion of a three band tight binding model, where large a_{1g}-e_g' level offset as well as third nearest neighbor hoppings between same orbitals is introduced in addition to the hoppings considered in ref.[10]. (b) Brillouin zone is shown in the extended zone scheme. The points denoted as SP are saddle points since the energy of the thick band at SP is lower than those around the gray circles, while it is higher than those around the open circles.

B. Method

To make the calculation feasible at low temperatures even in the presence of off-site repulsions up to second nearest neighbors, here we adopt a random phase approximation (RPA) approach. The singlet (V^s) and triplet (V^t) pairing interactions are given within RPA as follows.

$$V^s(\boldsymbol{q}) = U + V(\boldsymbol{q}) + \frac{3}{2}U^2\chi_s(\boldsymbol{q}) - \frac{1}{2}(U+2V(\boldsymbol{q}))^2\chi_c(\boldsymbol{q}) \quad (2)$$

$$V^t(\boldsymbol{q}) = V(\boldsymbol{q}) - \frac{1}{2}U^2\chi_s(\boldsymbol{q}) - \frac{1}{2}(U+2V(\boldsymbol{q}))^2\chi_c(\boldsymbol{q}) \quad (3)$$

Here, $V(\boldsymbol{q})$ is the Fourier transform of the off-site repulsions. χ_s and χ_c are the spin and charge susceptibilities, respectively, which are given as

$$\chi_s(\boldsymbol{q},\omega_l) = \frac{\chi_0(\boldsymbol{q})}{1-U\chi_0(\boldsymbol{q})}$$
$$\chi_c(\boldsymbol{q}) = \frac{\chi_0(\boldsymbol{q})}{1+(U+2V(\boldsymbol{q}))\chi_0(\boldsymbol{q})}. \quad (4)$$

Here χ_0 is the bare susceptibility given by

$$\chi_0(\boldsymbol{q}) = \frac{1}{N}\sum_{\boldsymbol{p}} \frac{f(\varepsilon_{\boldsymbol{p}+\boldsymbol{q}}) - f(\varepsilon_{\boldsymbol{p}})}{\varepsilon_{\boldsymbol{p}} - \varepsilon_{\boldsymbol{p}+\boldsymbol{q}}}$$

with $\varepsilon_{\boldsymbol{k}}$ being the energy dispersion measured from the chemical potential and $f(\varepsilon_{\boldsymbol{p}})$ is the Fermi distribution function. We take 128×128 k-point meshes in the actual numerical calculations.

To obtain the onset of the superconducting state, we solve the gap equation within the weak-coupling theory,

$$\lambda\Delta(\boldsymbol{k}) = -\sum_{\boldsymbol{k'}} V^{s,t}(\boldsymbol{k}-\boldsymbol{k'})\frac{\tanh(\beta\varepsilon_{\boldsymbol{k'}}/2)}{2\varepsilon_{\boldsymbol{k'}}}\Delta(\boldsymbol{k'}). \quad (5)$$

The transition temperature T_C is determined by the condition, $\lambda = 1$. In the present approach, ω dependence as well as the self energy correction is neglected. Although this approximation is quantitatively insufficient, we believe that the present approach suffices for the purpose of discussing the *competition* (relative tendency toward pairing) among various pairing symmetries.

C. Possible pairing symmetries

Pairing symmetries are classified according to the irreducible representations of the point group D_6, as shown in Fig.3. For the cases we considered, the f-wave pairing having B_2 symmetry turned out to have small values of λ, so in the following sections we consider s, p, d, f, i-wave pairings having A_1, E_1, E_2, B_1, A_2 symmetry, respectively.

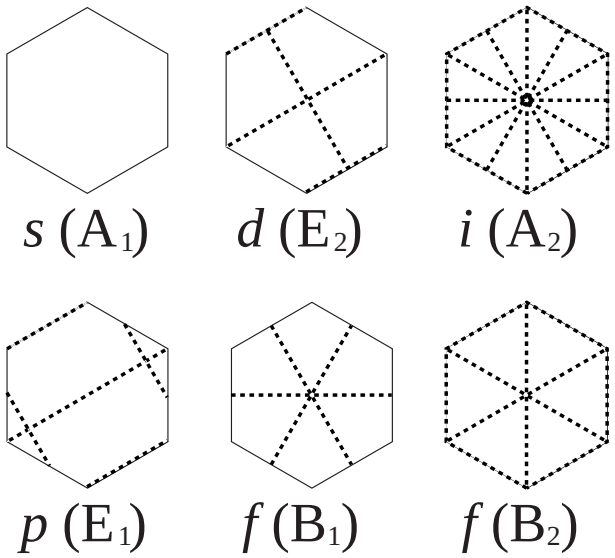

FIG. 3: Possible pairing symmetries of the present model classified according to the irreducible representations of D_6. The dashed lines represent the basic nodal lines.

III. RESULTS

A. Case without off-site repulsions

We first show results for a case without off-site repulsions, where we take $t_2 = t_3 = 0$, $t_4 = 0.18$, $n_h = 0.18$, $U = 2.4$ and $V_1 = V_2 = 0$. As seen in Fig.4, the f-wave pairing strongly dominates over other pairing symmetries, which is consistent with our previous study.[18] The reason for this is: (i) the spin fluctuations are nearly ferromagnetic (see Fig.8(a)), and (ii) the nodes of the f-wave gap do not intersect the Fermi surfaces as seen in Fig.5.

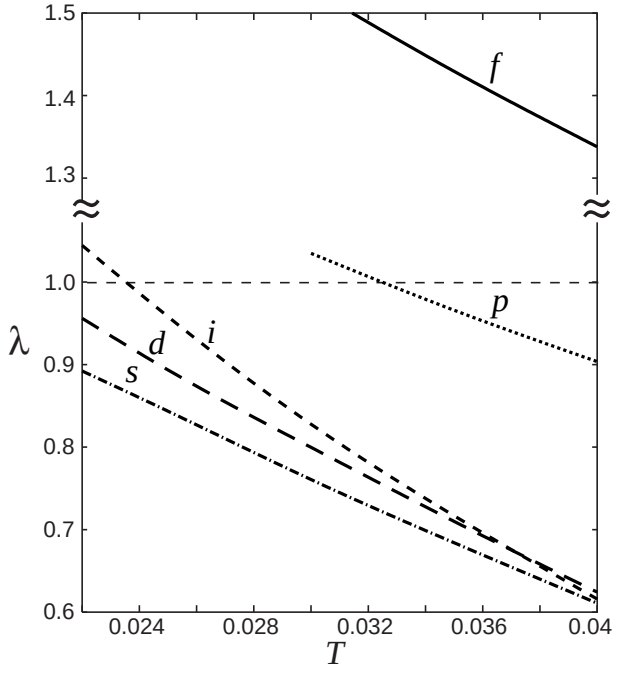

FIG. 4: The eigenvalue of the gap equation plotted as a function of temperature for each pairing symmetry. $t_2 = t_3 = 0$, $t_4 = 0.18$, $U = 2.4$, $V_1 = V_2 = 0$, $n_h = 0.18$.

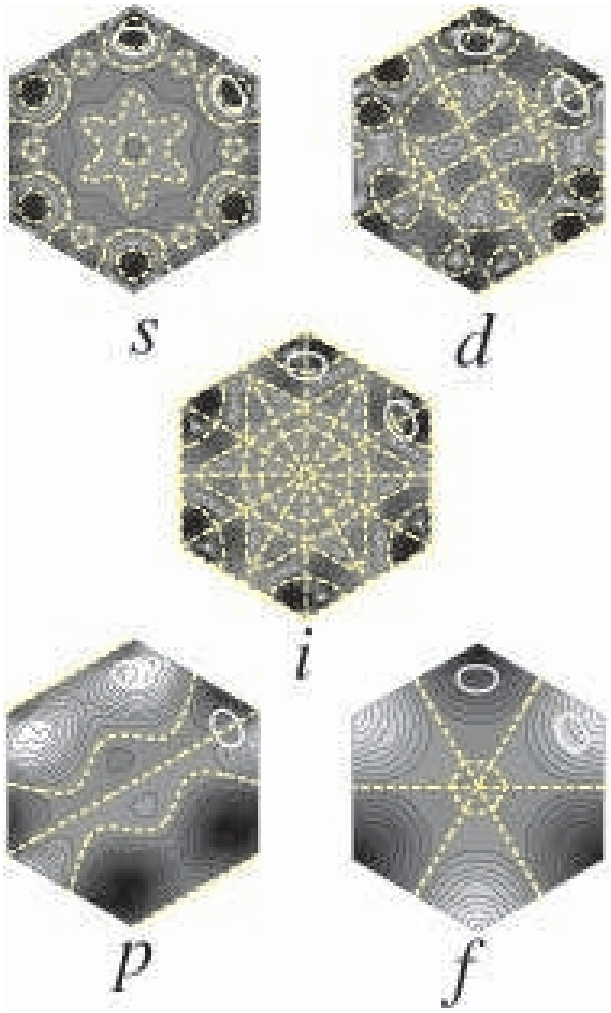

FIG. 5: Contour plot of the gap functions for each pairing symmetry. The dashed lines represent the nodes of the gap, and the white rings are the hole pockets (only two out of the six are shown for clarity). Parameter values are the same as in Fig.4 and $T = 0.03$. All the gap functions are plotted at $T = 0.03$ throughout the study.

B. Case with moderate nearest neighbor repulsion

We now turn on the off-site repulsions. As a case where a moderate nearest neighbor repulsion exists, we take $V_1 = 0.7$ and $V_2 = 0.4$, while other parameter values are taken to be the same as in section III A. We see in Fig.6 that the f-wave pairing still dominates over the others, but it is suppressed compared to the case without off-site repulsions. On the other hand, λ of singlet i,d, and s-wave pairings, along with the singlet gap functions shown in Fig.7, are barely affected by the off-site repulsions. To understand the reason why f-wave is suppressed while singlet pairings are unaffected by off-site repulsions, we show in Fig.8 the pairing interaction in momentum space. Here we define the "spin part" and the "charge part" of the pairing interaction as $U^2\chi_s(\boldsymbol{q})$ and $V(\boldsymbol{q}) - \frac{1}{2}(U + 2V(\mathbf{q}))^2\chi_c(\boldsymbol{q})$, respectively. The spin part peaks near the Γ point, while the charge part takes a broad negative minimum value around the M point. Thus, the effect of the pair scattering between two pockets that is mediated mainly by the charge part cancels out when there are nodes on the pockets (Fig.9(a)), while the sign change of the f-wave gap is unfavorable in the presence of the charge part, which has a negative sign (Fig.9(b)).

C. Case with large nearest neighbor repulsion

For a larger nearest neighbor repulsion, where we take the parameters values to be the same as in section III B

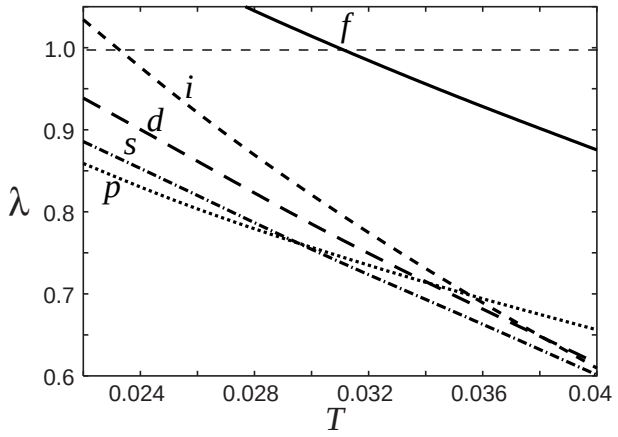

FIG. 6: A plot similar to Fig.4 with $V_1 = 0.7$, $V_2 = 0.4$, and other parameter values taken to be the same.

except that $V_1 = 1.0$, we find that f-wave pairing gives way to singlet pairings, as shown in Fig.10. Among the singlet pairings, which are found to closely compete in a wide range of parameter values, i-wave pairing surprisingly has the largest T_c for the present set of parameter values. Note that f-wave is no longer full-gaped on the Fermi surfaces due to the additional nodes as seen in Fig.11.

IV. DISCUSSION

A. Intuitive picture in real space

The reason why triplet pairings are suppressed by off-site repulsions while singlet pairings are barely affected can intuitively be understood in a real space picture. As seen in Fig.5 and Fig.7, the triplet pairing gap functions have relatively small number of nodes, which shows that these pairing takes place at short distances in real space. In particular, f-wave gap in the absence of off-site repulsions (Fig.5) essentially has the form $\sin(\frac{\sqrt{3}}{2}k_x+\frac{1}{2}k_y)-\sin(k_y)+\sin(-\frac{\sqrt{3}}{2}k_x+\frac{1}{2}k_y)$, which can be rewritten, apart from a constant, as

$$\begin{aligned} &\exp(i\boldsymbol{k}\cdot\boldsymbol{a}) - \exp(i\boldsymbol{k}\cdot\boldsymbol{b}) + \exp(i\boldsymbol{k}\cdot\boldsymbol{c}) \\ -\ &\exp(-i\boldsymbol{k}\cdot\boldsymbol{a}) + \exp(-i\boldsymbol{k}\cdot\boldsymbol{b}) - \exp(-i\boldsymbol{k}\cdot\boldsymbol{c}), \end{aligned} \quad (6)$$

where $\boldsymbol{a} = (\frac{\sqrt{3}}{2}, \frac{1}{2})$, $\boldsymbol{b} = (0, 1)$, $\boldsymbol{c} = (-\frac{\sqrt{3}}{2}, \frac{1}{2})$, and $\boldsymbol{k} = (k_x, k_y)$.

Since $\pm\boldsymbol{a}$, $\pm\boldsymbol{b}$, and $\pm\boldsymbol{c}$ are the coordinates (in units of the lattice constant) of the six nearest neighbors, the f-wave pairing in the absence of off-site repulsion is essentially a nearest neighbor odd-parity pairing. By contrast, the singlet pairings have many additional nodal structures compared to the basic nodal structure shown in Fig.3. This means that the pairs are mainly formed at large distances in real space. In fact, the result that λ is unaffected by V_1 and V_2 is consistent with the picture that the singlet pairs are formed at distances farther than second nearest neighbors.

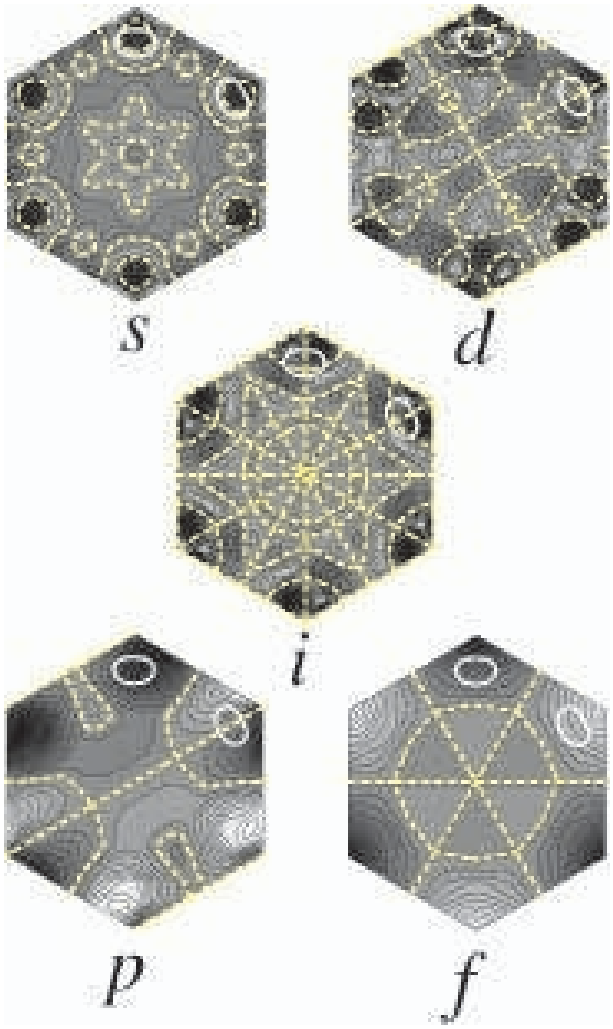

FIG. 7: Gap functions for the parameter values of Fig.6.

The reason why singlet pairs are formed at large distances while triplet pairs are formed at short distances can intuitively be understood from a nearly ferromagnetic spin alignment in real space as shown in Fig.12. Namely, electrons at close distances have nearly parallel spins, while the electrons can have antiparallel spins only when they are separated far away.

In order to reinforce this real space intuitive picture, we look into a case where the spin susceptibility peaks at positions more closer to the Γ point, namely when the spin structure is more purely ferromagnetic than in the cases considered in sections III B and III C. We take $t_4 = 0.15$ and $n_h = 0.1$, which moves the vHS and the Fermi level towards the band edge, thus making the spin structure more ferromagnetic as seen in Fig.13(a).

As shown in Fig.14(a), the triplet pairings dominate strongly over singlet pairings, and among the triplet pair-

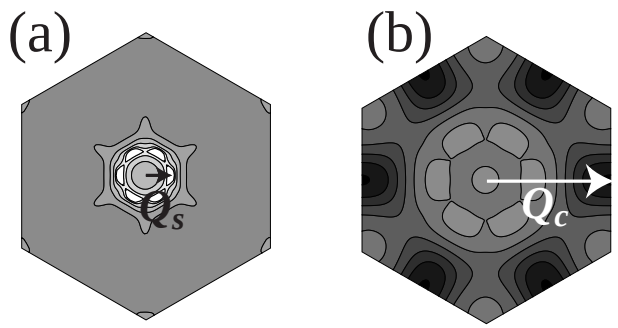

FIG. 8: Contour plots of the spin (a) and the charge (b) part of the pairing interactions for the parameter values of Fig.6. The spin part takes its maximum value ~ 25 at a short wave but finite vector $\boldsymbol{Q}_s$, while the charge part takes its minimum value ~ -3 at the M point.

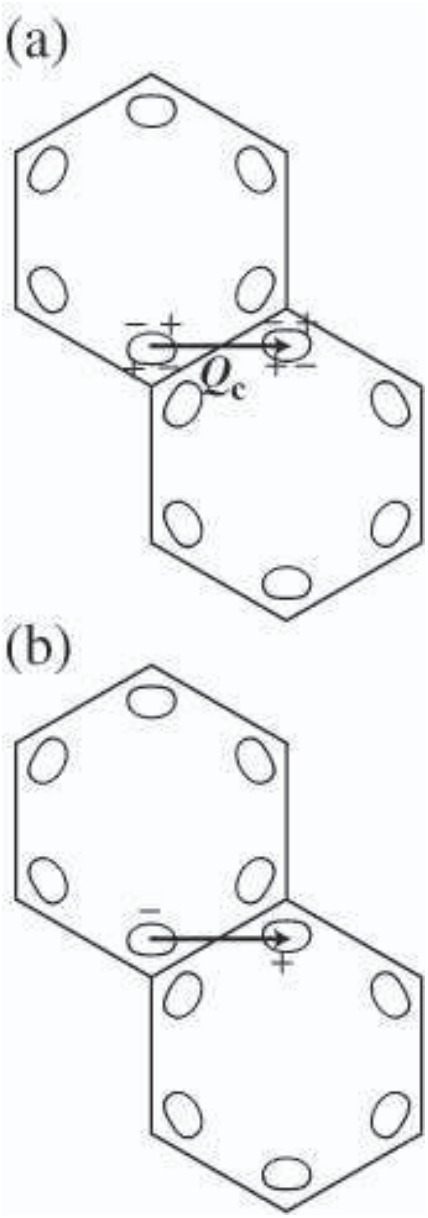

FIG. 9: The reason why singlet pairings (here we take the i-wave gap in Fig.7 as an example) are barely affected by off-site repulsions (a), while f-wave pairing is suppressed (b). Q_c is the wave vector at which the charge part of the pairing interaction takes its maximum absolute value.

ings, p-wave has the largest T_c. This result can again be understood from the above real space picture. Namely, since the spin susceptibility peaks around the Γ point, the wave length of the spin alignment is large, so that the singlet pairing distance is large from Fig.12. In fact, the number of nodes in the singlet pairing gaps are found to increase compared to those in sectionIII C, as seen in Fig.15. This makes the singlet pairing less competitive against triplet pairings. Moreover, since the wave length of the spin alignment is large, not only electrons residing at nearest neighbors but also those separated to some extent have parallel spins, so that spin triplet pairing can take place at relatively large distances, thereby circumventing the effect of off-site repulsions. It can be seen from Fig.15 that the f-wave gap has additional nodes close to the Fermi surfaces suggesting that the pairing takes place not only at nearest neighbors but also at larger distances. Pairing at large distances, and thus the presence of nodes near the Fermi surfaces, degrades f-wave pairing, so that p-wave pairing dominates.

Even if we decrease V_1 down to $V_1 = 0.7$, p-wave still dominates over f-wave as seen Fig.14(b), in contrast to the case discussed in sections III B. This is because in the presence of purely ferromagnetic spin fluctuations, f-wave pairing takes place at large distances even for relatively small off-site repulsions, so that the additional nodes are located close to the Fermi surfaces. This can in fact be seen by comparing Fig.15 and Fig.16, where the additional nodes in the f-wave gap do not move away so much from the Fermi surfaces even if we reduce V_1 from 1.0 to 0.7. This is in contrast with the case of Fig.7 and Fig.12, where the additional nodes in the f-wave gap clearly moves away from the Fermi surfaces (i.e., the nearest neighbor pairing component becomes large) as we reduce V_1.

B. Peculiar Disconnectivity of the Fermi surfaces

The reason why i-wave pairing, which has a very large angular momentum, may dominate in the presence of off-site repulsions can be found in the disconnectivity of the Fermi surface *peculiar* to the present system. Namely, the Fermi surfaces in the present case are disconnected in a way that they do not intersect the Γ-M lines nor the hexagonal Brillouin zone edge, both of which are the nodal lines of the i-wave gap. Thus, the number of nodal

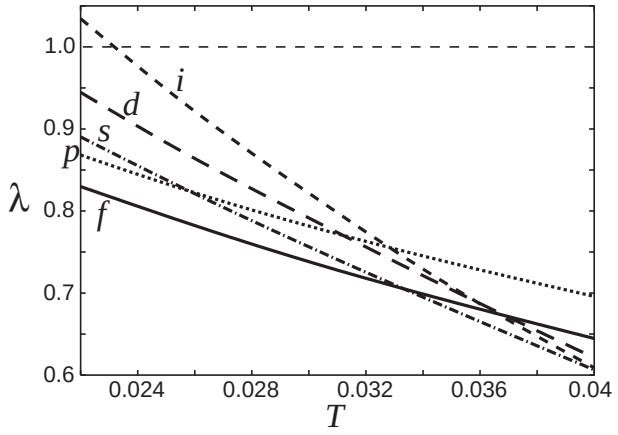

FIG. 10: A plot similar to Fig.6 with $V_1 = 1.0$ and other parameter values taken to be the same.

lines that intersect the Fermi surfaces is similar among the three singlet pairing symmetries, extended s, d, and i-waves, as seen in Fig.11, thereby making the competition subtle. In fact, we have looked into various sets of parameter values and found that the three singlet symmetries tend to have close values of λ.

To show more clearly that the subtle competition between i-wave and other singlet pairings is due to the disconnectivity of the Fermi surfaces peculiar to the present system, we compare the above results with those for cases with only nearest neighbor hopping, namely with a simple triangular lattice. In a simple triangular lattice with $n_h < 0.5$, the Fermi surfaces are disconnected in a manner that they do not intersect the Γ-M lines, but they do intersect the Brillouin zone edge. The absence of t_4 moves the van Hove singularity away from the band top, so in order to have a moderate T_c as in the cases above, we either have to take larger values of n_h, or increase the values of the electron-electron interactions. Here we take $n_h = 0.4$ and the interactions the same as in sectionIII C in Fig.17(a), while in Fig.17(b) we take the same n_h but take the interactions 1.5 times larger than in sectionIII C. It can be seen that in both cases, λ of the i-wave pairing is much smaller than that of the d- and the s-wave pairings. This is because the Fermi surfaces intersect the Brillouin zone edge, i.e., the nodes of the i-wave gap, as seen in Fig.18 and Fig.19.

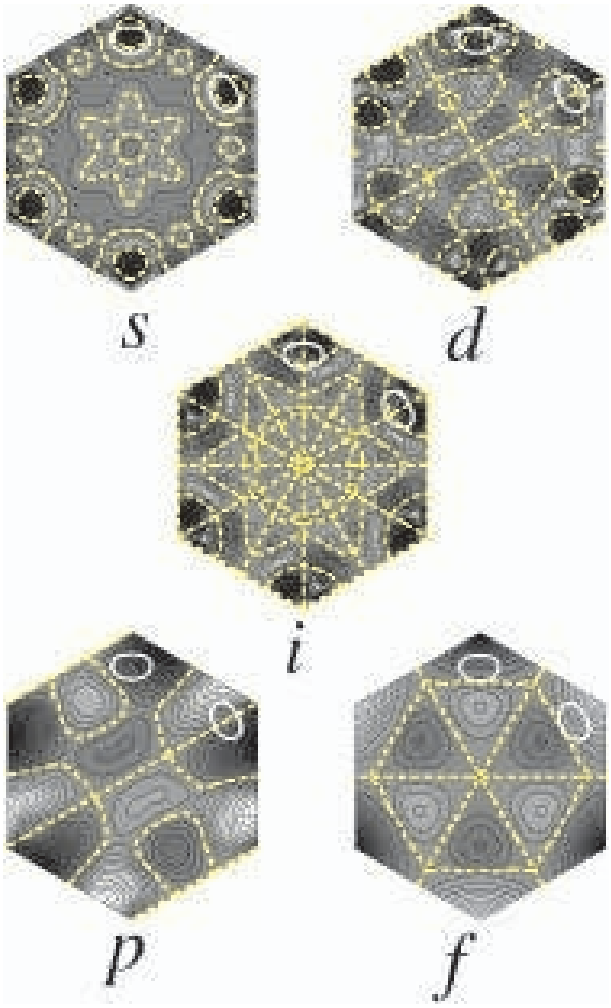

FIG. 11: Gap functions for the parameter values of Fig.10.

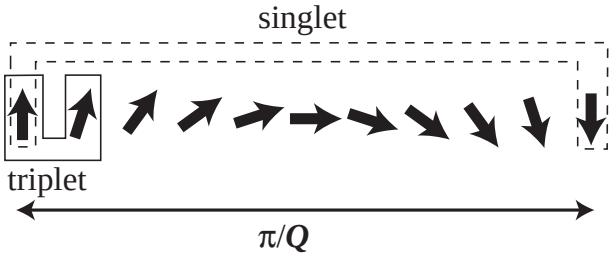

FIG. 12: An intuitive real space picture of singlet and triplet pairings when nearly ferromagnetic spin fluctuations are present.

C. Comparison with the case with antiferromagnetic spin fluctuations

In the present study, we have looked into a model where nearly ferromagnetic spin fluctuations and charge fluctuations due to off-site repulsions coexist. It is interesting to compare the present situation with the case where *antiferromagnetic* spin fluctuations and charge fluctuations coexist. We have previously proposed that spin-triplet pairing is possible in a quarter-filled, quasi-1D organic material $(TMTSF)_2X$,[36,37] where superconductivity lies right next to spin density wave (SDW) in the pressure-temperature phase diagram, so that the spin fluctuations should be antiferromagnetic. Naively, antiferromagnetic spin fluctuations would favor spin singlet d-wave pairing, while experiments suggest triplet pairing.[39,40] Our proposal is that if $2k_F$ ($= \pi/2$ because

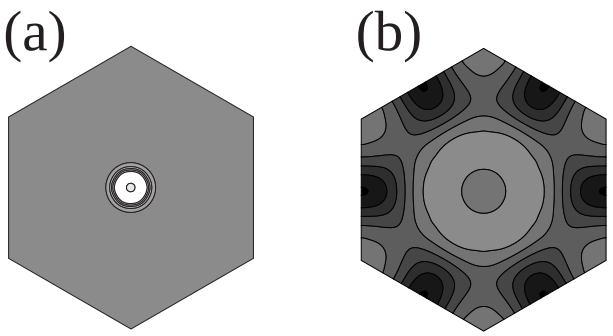

FIG. 13: The spin (a) and charge (b) part of the pairing interaction for $t_2 = t_3 = 0$, $t_4 = -0.15$, $U = 2.4$, $V_1 = 1.0$, $V_2 = 0.4$, $n_n = 0.1$. The spin part takes its maximum value ~ 35 at a vector $\boldsymbol{Q}_s \sim \mathbf{0}$, while the charge part takes its minimum value ~ -3 at the M point.

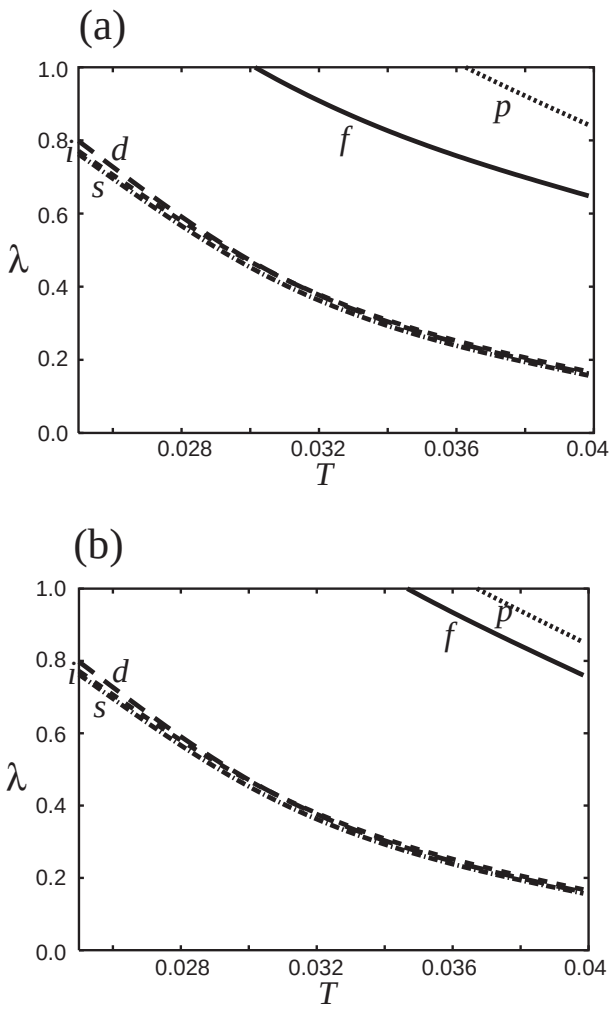

FIG. 14: Plots similar to Fig.6 for $t_2 = t_3 = 0$, $t_4 = 0.15$, $U = 2.4$, $V_2 = 0.4$, $n_n = 0.1$. $V_1 = 1.0$ (a) and $V_1 = 0.7$(b).

the band is quarter-filled) charge fluctuations coexist with $2k_F$ spin fluctuations, as suggested from an experimental fact that CDW actually coexist with SDW[41,42], spin triplet 'f-wave' like pairing, which is essentially a fourth neighbor pairing having a gap form of $\sin(4k_x)$ (this is called f-wave in the sense that the gap changes sign as $+-+-+-$ along the Fermi surface), may become competitive against singlet 'd-wave' like pairing, which is a second neighbor pairing with a gap form $\cos(2k_x)$. In momentum space, this close competition arises because (i) the number of nodes of the f-wave gap on the Fermi surface is the same with that of the d-wave because of the disconnectivity of the Fermi surfaces due to quasi-one-dimensionality, and (ii) the pairing interactions due to spin and charge fluctuations (apart from the first order terms) have the form $V^s = \frac{3}{2}V_{\rm sp} - \frac{1}{2}V_{\rm ch}$ for singlet pairing and $V^t = -\frac{1}{2}V_{\rm sp} - \frac{1}{2}V_{\rm ch}$ for triplet pairing, where $V_{\rm sp}$ and $V_{\rm ch}$ are contributions from spin and charge fluctuations, respectively, so that the absolute values of V_s and V_t becomes comparable when $V_{\rm sp} \simeq V_{\rm ch}$. A similar theory has been proposed by Fuseya *et al.*[38] Recently, we have confirmed this scenario microscopically by applying RPA to an extended Hubbard model with off-site repulsions up to third nearest neighbors.[43] Here, a key parameter is the second nearest neighbor repulsion, due to which the $2k_F$ charge fluctuations arises.

In real space, this f-d competition can be understood as follows. When there are purely antiferromagnetic spin fluctuations, second neighbor singlet pairing is likely to occur, as seen from Fig.20. If we now turn on the second nearest neighbor repulsion, the second neighbor singlet pairing is degraded, and a fourth neighbor triplet pairing becomes competitive. Since this intuitive picture in real space is rather general, the tendency that spin singlet pairing, which is favored by antiferromagnetic spin fluctuations, gives way to triplet pairings in the presence of sufficiently large off-site repulsions, should hold in various situations. In fact, such a singlet-triplet crossover due to off-site repulsions is seen also in a half-filled quasi-1D system,[48] a model for Sr_2RuO_4 on a 2D square lattice,[49] and a model for Na_xCoO_2 on a 2D triangular lattice.[17]

Now, it is interesting to point out that the above tendency is exactly the opposite of what has happened in the present case with nearly ferromagnetic spin fluctuations : triplet pairings formed at relatively short distances give way to singlet pairings formed at farther distances when sufficiently large off-site repulsions are introduced. Thus, the effect of the off-site repulsion totally differs depending on whether the spin fluctuations are ferromagnetic or antiferromagnetic. It is also important to note that in both cases, the disconnectivity of the Fermi surfaces can assist the pairings at large distances (singlet in the ferromagnetic case and triplet in the antiferromagnetic case), which have larger number of nodes in the gap function.

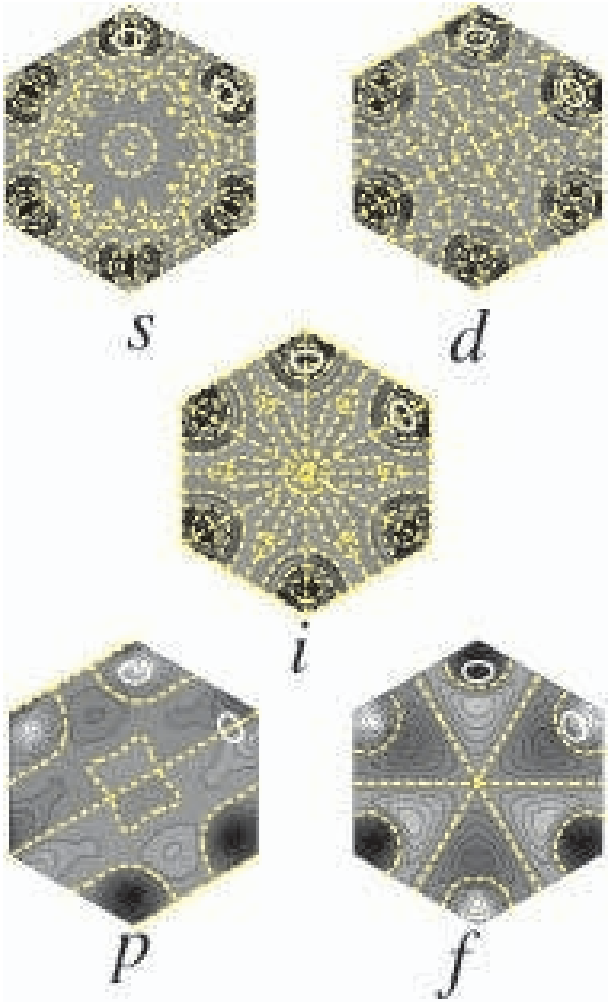

FIG. 15: Gap functions for the parameter values of Fig.14(a).

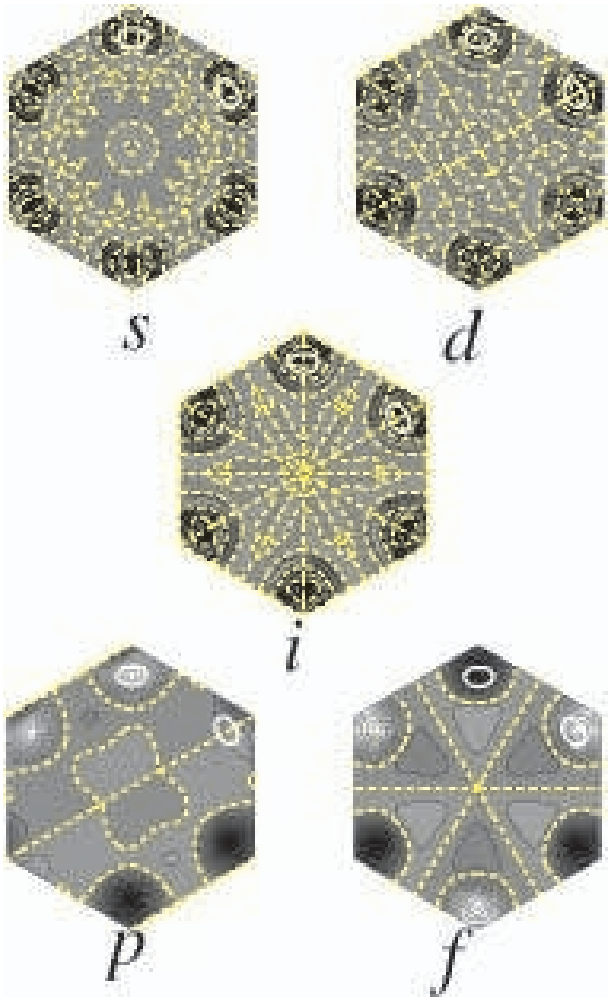

FIG. 16: Gap functions for the parameter values of Fig.14(b).

D. Validity of focusing on the pockets

In the present study, we have focused only on the e'_g Fermi pockets. Although it is necessary to consider a multiband model[10] in order to strictly take into account the effect of the Fermi pockets, our single band approach is supported by a recent multiband FLEX study,[19] where a similar f-wave pairing is found to dominate when spin fluctuations are not too purely ferromagnetic.

Experimentally, the Fermi pockets are not observed in ARPES experiments[44,45] up to date. However, since these experiments are done for materials with relatively large Na content, (i.e., large number of electrons in CoO layers) it is likely that the Fermi level lies above the e'_g bands.[46] In fact, an experimental result that maximum T_c is reached only when the content of Na decreases sufficiently[47] can be considered as an indirect support for scenario in which the e'_g band plays an important role for superconductivity.

E. Pairing symmetry in the cobaltate

Let us finally discuss the pairing symmetry in the cobaltate in light of the present results along with the existing experiments. We have seen that "full-gapped" f-wave proposed in ref.18 is robust to some extent even in the presence of off-site repulsions. Existence of nodes in the gap function is suggested in a number of experiments,[4,5] which may seem to contradict with the f-wave scenario. In the actual Na_xCoO_2, however, there is also the large Fermi surface around the Γ point, which is not taken into account in the present study. Then we can propose a possible scenario where the 'active' Fermi surfaces for superconductivity are the pockets, while the large Fermi surface is a 'passive' one, in which superconductivity is induced by the 'active' band via interband interactions. Since the nodes of the f-wave gap intersects the large Fermi surface, this can account for the experimental results suggesting the existence of gap nodes. In fact, it has been shown that a μSR data can be explained

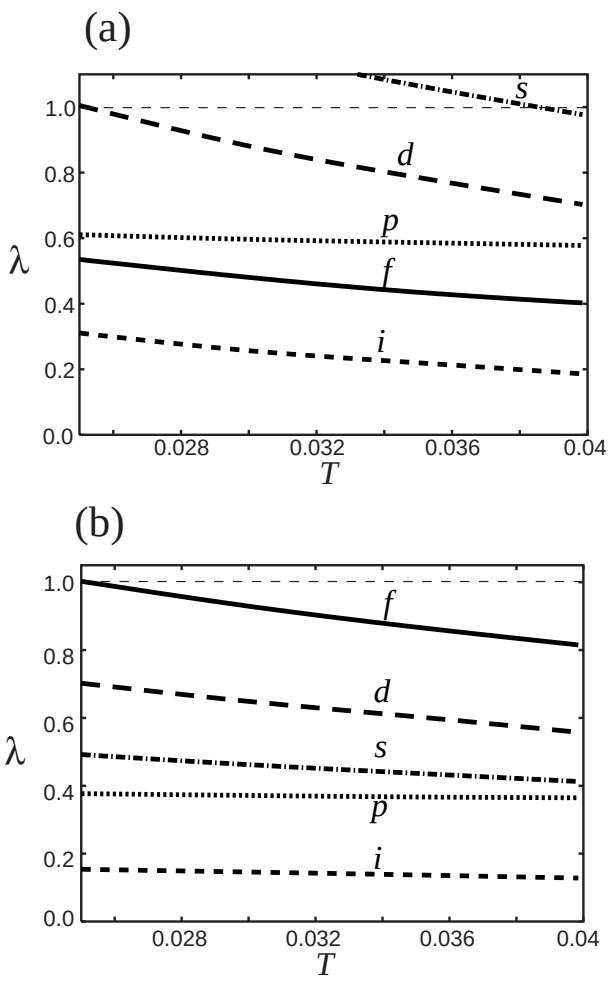

FIG. 17: Plots similar to Fig.6 for $t_2 = t_3 = t_4 = 0$. $U = 2.4$, $V_1 = 1.0$, $V_2 = 0.4$, and $n_h = 0.4$ (a), $U = 3.6$, $V_1 = 1.5$, $V_2 = 0.6$, and $n_h = 0.18$ (b).

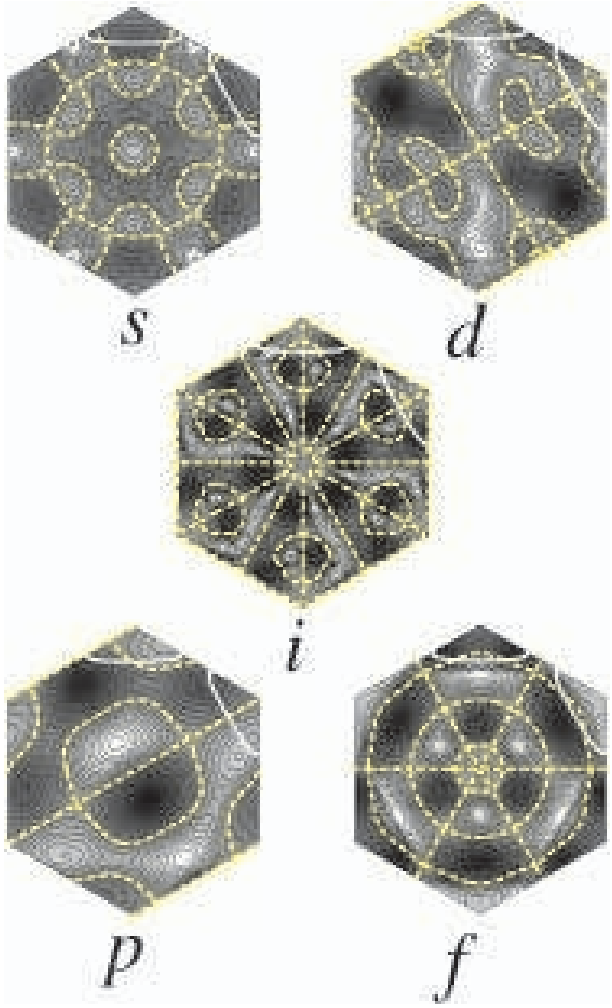

FIG. 18: Gap functions for the parameter values of Fig.17(a).

by taking this view.[9]

On the other hand, singlet pairings are found to dominate over triplet pairings in the present model when spin fluctuations are not purely ferromagnetic and the off-site repulsions are sufficiently large. Among the singlet symmetries, i-wave is another good candidate that competes with f-wave since it does not break time reversal symmetry below T_c. We have shown that i-wave may dominate over other symmetries, but the parameter range where this symmetry dominates is not so wide as that for the f-wave. Although extended s-wave is also consistent with almost all the existing experiments, we could not find a parameter regime where this symmetry dominates. However, considering the fact that the competition within the singlet pairings is very subtle, and that the present model only roughly takes into account the electronic structure of the actual material, we believe the possibility of extended s-wave pairing cannot be ruled out.

Since d-wave and p-wave pairings are likely to result in a broken time reversal symmetry and/or a full gapped state (unless an accidental situation occurs) below T_c due to the two-fold degeneracy, we believe that the possibility of these pairing states are less likely. We may conversely say from the present results that the spin fluctuation in the actual cobaltate is not *purely* ferromagnetic and/or the off-site repulsions are not too large, so that p-wave pairing does not dominate.

At present, it is not clear experimentally whether the pairing occurs in the singlet or in the triplet channel because Knight shift experiments are done on polycrystal samples. In order to settle this singlet-triplet debate, Knight shift experiment for high quality samples, or alternatively, tunneling spectroscopy studies based on a newly developed theory for triplet superconductors proposed by one of the present authors[50] is required. Thus, our viewpoint is that possibility of neither singlet nor triplet pairings cannot be ruled out experimentally at the present stage. Then, the bottom line obtained from the present study, combined with the existing experimental

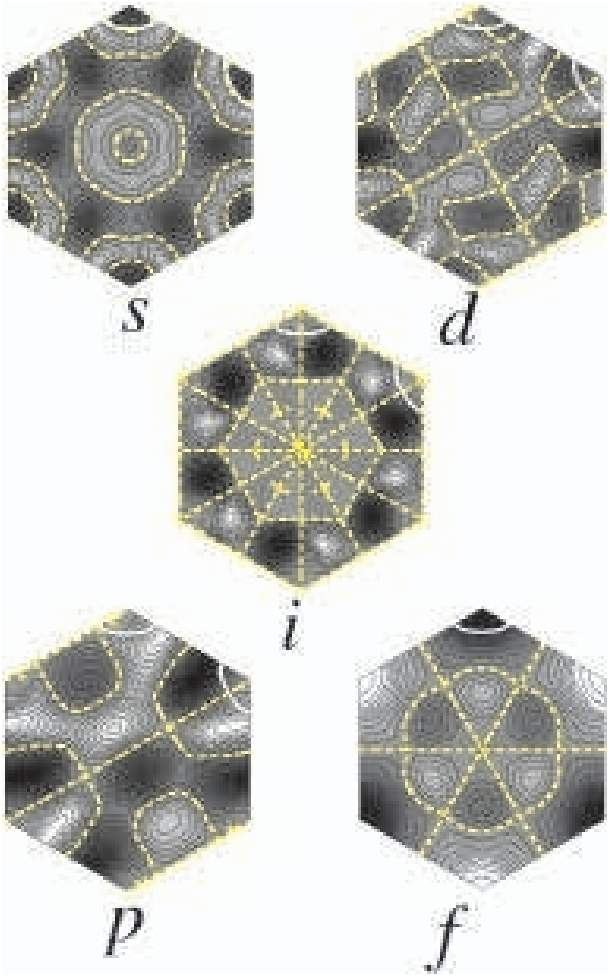

FIG. 19: Gap functions for the parameter values of Fig.17(b).

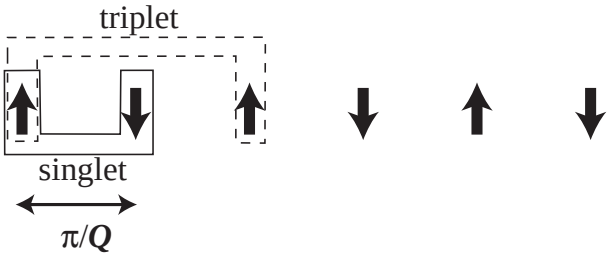

FIG. 20: An intuitive real space picture of singlet and triplet pairings when antiferromagnetic spin fluctuations are present. Note that when $Q = 2k_F = \pi/2$ as in the case of $(TMTSF)_2X$, arrows in this figure are separated by two lattice spacings.

facts, is that triplet f-wave pairing is most likely, although there remains a possibility of singlet i-wave pairing, and also a possibility of extended s-wave pairing cannot be ruled out. This ambiguity is a direct consequence of the very point of the present study. Namely, the present system is highly peculiar in that anisotropic pairings having large angular momentum, such as f- and i- waves, are good candidates due to the disconnectivity of the Fermi surfaces. The fact that these "unusual" pairing symmetries are good candidates makes the pairing competition subtle.

V. SUMMARY

To summarize, we have studied an effective single band model for the hydrated sodium cobaltate focusing on the e'_g hole pockets, where we take into account off-site repulsions up to second nearest neighbors. We find that f-wave pairing proposed in our previous study is robust to some extent in the presence of off-site repulsions. For large off-site repulsions, f-wave gives way to singlet pairings that does not break time reversal symmetry below T_c.

Acknowledgments

Calculation has been performed at the facilities of the Supercomputer Center, Institute for Solid State Physics, University of Tokyo

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